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HEI Workshop on Fuel Composition and PM – December 8, 2016
Executive Summary[1]

On December 8, 2016, the Health Effects Institute (HEI) hosted a workshop in Chicago on the "Effects of Fuel Composition on PM," attended by approximately 45 participants representing regulatory bodies, the automobile and fuel industries, academic and governmental research organizations, and trade organizations. The workshop was prompted, in part, by uncertainties regarding particulate matter (PM) emissions from gasoline-fueled vehicles, and the complex effects that fuel composition and engine technology changes appear to have on such PM emissions.

The main goal of the workshop was to present and summarize the current state of knowledge on the effects of fuel composition on PM – including both primary emissions and secondary formation. While some information was included about diesel fuels and vehicles, the primary emphasis was on gasoline applications. A broad set of questions was posed to help set the stage for the workshop and to guide further discussions. The questions included:

- What is the relationship between emission tests, including the effects of fuel reformulation, under laboratory/certification conditions and real-world emissions and exposures?
- What is the likely impact/benefit in terms of human exposure from: (1) ethanol blending, (2) fuel aromatics content, and (3) the use of gasoline direct injection (GDI)? What do we know and what are the research needs?
- Is GDI a game changer: Will it change the picture of human exposure, especially when taken in the context of fuel formulations and ethanol blending?
- What are some of the challenges in meeting the US Tier 3 and California Low Emission Vehicle 3 (LEV3) standards? What is the role of ethanol, or more generally fuel formulation, in this context?

This summary draws from the workshop presentations and highlights the topics that were discussed. *The workshop agenda is attached and the presentations are available on the HEI website* *here*.

Regulations, Engines, Aftertreatment, and Fuels

- PM emissions from modern vehicles are very low, making reliable measurements challenging.
- Vehicle PM emission standards are becoming increasingly stringent. In the United States, for light duty vehicles (LDVs), the PM mass emission limit under the Tier 3 standard is 3 mg/mi (to be phased

[1] Published on September 12, 2017, by the Health Effects Institute.

IMPORTANT MESSAGES FROM THE WORKSHOP

- Biofuels and new automotive technologies have been introduced in response to concerns about several issues, especially climate change; however, the impact of these introductions on emissions and human exposure is not well understood. This workshop was organized to gain some insights into these important questions.
- Biofuel policies and usage are evolving in both the US and Europe. At present, the US Renewable Fuel Standard ensures that nearly all gasoline contains 10% ethanol (E10). On the technology side, the gasoline direct injection system is being widely adopted for the light duty fleet.
- The results of laboratory testing show that the effects of ethanol blending on particulate matter (PM) emissions – particularly for low-blend gasoline (E10 – E20) – are variable, reflecting the complex and competing effects that ethanol has on in-cylinder PM formation.
- Real-world test findings also highlight that vehicle deterioration may have a much larger effect on fleet-wide emissions than do fuel modifications. And, laboratory findings highlight the importance of including secondary organic aerosol levels when considering human exposures as these may exceed tail-pipe emissions tenfold or greater.
- There continues to be an important need to better understand the impact of fuel composition — including ethanol blending and technology — on PM, air toxics and other emissions, taking into account the various fuels, vehicle, and engine parameters. Also important is a better understanding of the atmospheric transformations of the various pollutants and their impact on human exposure.

in between 2017-2022); California standards specify a 1 mg/mi limit (to be implemented in 2025). The US has no particle number (PN) standard.

- European PM mass standards for light-duty vehicles are slightly less stringent than those in the U.S., but the Euro 6 standard also includes a limit on PN that is effectively more stringent in controlling PM emissions than the US mass standard. The interim gasoline direct injection vehicle PN standard of 6×10^{12} is being reduced in 2017 by an order of magnitude to 6×10^{11}, the same level as that for diesel vehicles. However, the measurement method used in Europe to quantify PN likely underestimates the total PN emissions, because it does not account for particles smaller than 23 nm or volatile aerosols. Europe has also recently introduced a Real Driving Emission certification testing requirement.
- Motivated by climate change, energy security, and other concerns, most major vehicle markets now have some form of fuel economy standard for LDVs, and several countries (including the US, China, and Japan) also have standards for heavy-duty vehicles (HDVs). There are currently no HDV fuel economy standards in Europe, though this is being actively discussed.
- Biofuel policies and usage are evolving in both the US and Europe. At present, the US Renewable Fuel Standard ensures that nearly all gasoline contains 10% ethanol (E10). The use of higher blend levels of ethanol in the US has been challenging and controversial.
- Ethanol has a higher octane value than gasoline; higher blends of ethanol — depending on other components of the gasoline — have the potential to create higher octane fuels, which allow for engines to operate with a higher compression ratio and improved fuel efficiency.

- GDI technology is rapidly being deployed throughout the LDV fleet because it offers engine performance and fuel efficiency benefits. GDI enables higher compression ratios and is often used in conjunction with turbo-charging, variable valve timing, and engine downsizing, all of which help improve vehicle efficiency.
- Compared to traditional port fuel injection (PFI) engines, GDI engines produce higher PM and PN emissions under typical testing conditions; some studies have also reported higher emissions of polycyclic aromatic hydrocarbons (PAHs).
- Emissions of total non-methane hydrocarbons and the organic carbon fraction of PM from pre-LEV, LEV1, LEV2, and SULEV vehicles decrease with the newer technology vehicles. However, the elemental carbon fraction does not follow this pattern and the GDI vehicles have higher elemental carbon emission factors than the older PFI vehicles.
- The California Air Resources Board has deployed a suite of toxicity screening assays to assess oxidative stress, inflammation, and mutagenicity effects of vehicle emissions. Application to PM emissions from various vehicle types has shown that toxicity of PM from old diesel technology vehicles is far higher than from modern gasoline or diesel vehicles when expressed on a per-mile basis, reflecting the much higher PM emissions from old technology diesel engines. However, when expressed on a per-mass basis, PM toxicities from the various vehicles other than the old technology diesel vehicle are relatively similar and it is difficult to discern a pattern among the observed differences.

Laboratory Tests of Gasoline Engine Emissions

- In 2010, scientists from Honda published results of a study that shows that both PN and PM emissions from gasoline engines can be predicted by the PM index (PMI), which is calculated based on the weight fraction, vapor pressure, and double-bond-equivalent value of each gasoline constituent.
- In the federally funded EPAct/V2/E-89 study, the largest such study to-date, the effects of multiple fuel properties on PM emissions from a fleet of low-mileage, Tier 2, PFI gasoline engines were investigated. The fuels used in these studies were 'match blended.'[2] In 10 of the 15 vehicles examined, PMI was an excellent predictor of PM emission rates. In addition, the fuel ethanol content (from E0 to E20) had a 'reinforcing effect' on PM, resulting in higher PM emissions. EPA hypothesized that this effect is the result of combustion chamber cooling due to ethanol's high heat of vaporization, which enhances the formation of PM from low volatility gasoline components. However, for the other five vehicles examined, the fuel PMI and ethanol were not well correlated with PM mass emissions, suggesting that vehicle-specific factors are also important in determining the effects of fuel composition on PM emissions.
- Several other studies of the effects of fuel blending on vehicle emissions, though smaller in scope than the EPAct program, have examined the effects of similar ethanol content but have used 'splash

[2] Match blending is the process by which the base fuel is altered when ethanol is added – for example, by adding BTEX (benzene, toluene, ethylbenzene and xylene) – so that the other fuel properties (e.g. effective octane rating, boiling point, etc.) can be made to match desired specifications; this allows for greater control over properties of the blended fuel.

blended'[3] fuels. Some of these studies have found that PM and PN emissions are reduced with increasing levels of ethanol (or iso-butanol); these authors have attributed this reduction to ethanol disrupting the chemistry that forms soot from precursor compounds. These studies also report that PM and PN emissions increase with gasoline aromatics content, as predicted by the PMI equation.

- Overall, the results of laboratory testing show that the effects of ethanol blending — particularly for low-blend gasoline (E10 – E20) — on PM emissions are variable, reflecting the complex and competing effects that ethanol has on in-cylinder PM formation. The published data suggest that ethanol can both suppress (by disrupting soot formation chemistry) and enhance (by reducing in-cylinder temperature) PM formation.
- The relative importance of the different mechanisms depends on details such as fuel composition, engine design, and engine calibration. For example, the factors influencing test results include:
    - fuel properties (hydrocarbon composition and PMI [levels of unsaturated hydrocarbons and aromatic content] and various physical parameters [T_{50}, T_{90} and Research Octane Number] as well as how the blends are prepared);
    - engine operating conditions and the test cycle used;
    - GDI configuration; and
    - engine type (including the manufacturer, model and engine design), to name just a few.

  Given such complexities and the expense of testing, relatively few studies have been performed to systematically parse the effects of these different factors in detail. Results of the studies published so far have been inconsistent and their interpretation disputed.
- While PN emission rates have been found generally to correlate with the fuel's PMI values in tests that included the entire federal test procedure (FTP) cycle, they are much higher during the cold-start cranking portion of the FTP (i.e., the first 25 seconds) and during high acceleration, suggesting that the competition between particle formation and oxidation processes can vary under different operating conditions. PM emissions are also higher under low temperature (i.e., between -7 and -18 °C) and under engine start-stop driving conditions.
- There are two configurations of GDI technology in the market: wall-guided and spray-guided injection systems. Early testing from a limited number of vehicles suggests higher PM emissions from wall-guided compared to spray-guided GDI engines. However, the technology is evolving rapidly as manufacturers optimize designs and engine calibrations. Newer GDIs incorporate features of both configurations; they generally have lower PM emissions than first-generation engines, and some can meet the stringent US Tier 3 standard (3 mg/mi).
- This is an active area of research and additional studies have been published since the workshop.[4] Also, other studies are underway with the objective of clarifying the effects of fuel properties on

[3] In splash blending, ethanol is added to the base fuel volumetrically to produce the desired percent blend; the other properties of the blend are generally altered in this process as a result of dilution and other interactions. Currently, most commercial gasoline in the US is splash blended.

[4] For example, see: Coordinating Research Council. 2017. Evaluation and Investigation of Fuel Effects on Gaseous and Particulate Emissions on SIDI In-use Vehicles. CRC, Alpharetta, GA and Sakia, S, and Rothamer, D. 2017. Effect of ethanol blending on particulate formation from premixed combustion in spark-ignition engines. *Fuel*, 196: 154-168.

emissions from GDI vehicles. For example, the Coordinating Research Council and the University of California at Riverside are engaged in such work. Through the Co-Optima program, the US Department of Energy is coordinating a very comprehensive new effort to optimize fuels and engines together as a system, thereby enabling future vehicles with improved performance, efficiency, and sustainability.

Linking Tailpipe Emissions to Real-World Measurements

- Automobile exhaust comprises a complex mixture of gases and particles. In the atmosphere, the emissions react to form secondary PM — both secondary organic aerosols (SOA) and nitrate. Primary emissions and secondary PM formation are both important to consider when assessing human exposure to traffic-related air pollution. Primary PM emissions likely dominate human exposure in the near-road environment; secondary PM likely dominates exposure as one moves away from roads.
- Direct measurements show that during the last several decades tail-pipe emissions of exhaust components from both LDV and HDVs have gone down, in many cases by an order of magnitude or more. This has led to improved overall air quality in most parts of the United States. For example, ambient measurements in Southern California have shown that organic aerosol concentrations have been reduced from 15 to 5 $\mu g/m^3$ (annual average) over the past 40 years, with most of this reduction attributed to lower SOA precursor emissions from LD vehicles.
- In a roadway tunnel study, primary organic aerosol (POA) emissions were dominated by lube oil constituents, regardless of the mix of LDVs versus HDVs in the traffic. It seems therefore likely that high POA emissions represent an in-use problem for a small number of vehicles and are probably caused by engine wear or vehicle deterioration, and that POA mass emissions from the in-use vehicle fleet may not be greatly affected by changes in fuel composition. In comparison, fuel composition plays a more important role in black carbon (BC) emissions and in SOA formation and therefore can affect ambient air quality and human exposure.
- These findings also highlight that vehicle deterioration may have a much larger effect on fleet-wide emissions than do fuel modifications.
- Tailpipe emissions introduced into a smog chamber and then subjected to various conditions provide a good way to study SOA formation. When exposed to oxidative conditions that simulate ambient conditions in a chamber study, SOA and nitrate form rapidly. Secondary PM formation peaks after about 2-days of oxidation, when the amount of SOA exceed the amount of POA by 1 to 2 orders of magnitude. These observations highlight the importance of including SOA levels when considering human exposures; they also point to the importance of controlling PM precursors as a way to reduce total exposure.
- For *gasoline* exhaust, SOA formation is strongly dependent on the aromatic content of the fuel and there appears to be no difference between SOA production from PFI vs GDI vehicles. In contrast, for *diesel* vehicles SOA formation does not show any clear fuel composition effect.
- Overall, the amount of SOA formation from LDV emissions has been reduced with newer technology vehicles, including both PFI and GDI vehicles.

Future challenges and Research Needs

There continues to be an important need to better understand the impact of fuel composition — including ethanol blending, and technology — on PM, air toxics, and other emissions, taking into account the various fuels, vehicle, and engine parameters. The workshop highlighted the need for further research on:

- The impacts of advanced engine technologies (e.g., GDI and turbocharging) and emissions controls (e.g., gasoline particulate filters) on PM formation, and how this may be affected by fuel formulation;
- The effects of fuel formulation on the complex chemistry involved with direct PM emissions, as well as on SOA formation in the atmosphere; and
- The contributions of gasoline and diesel on-road vehicles to primary and secondary PM emissions and ambient PM, and their impact on human exposure.

WORKSHOP ON EFFECTS OF FUEL COMPOSITION ON PM[5]

December 8, 2016

Renaissance Chicago O'Hare Suites (Salon AB), Chicago

AGENDA

Workshop presentations available here

7:30 am **Breakfast (Salon D)**

INTRODUCTION AND BACKGROUND

Chair: David Foster, University of Wisconsin

8:30 am Welcome and introduction to the workshop, *Daniel Greenbaum, Health Effects Institute*, and *Allen Robinson, Carnegie Mellon University*

8:50 am Policy background and future directions in the US and Europe, *John German, International Council on Clean Transportation*

9:20 am The gasoline engine as a system and the role of new technology, *Timothy Johnson, Corning*

9:50 am Gasoline fuel – Formulation issues and constraints, and background to emissions testing, *John Farrell, National Renewable Energy Laboratory*

10:20 am **Break**

ATMOSPHERIC AND AMBIENT PM

Chair: Rashid Shaikh, HEI

10:40 am Trends and sources of primary PM emissions from gasoline and diesel vehicles, *Robert Harley, University of California at Berkeley*

11:10 am Linking tailpipe to ambient – primary emissions versus secondary PM formation, *Allen Robinson, Carnegie Mellon University*

FUEL COMPOSITION, ENGINE TECHNOLOGY, AND PM EMISSIONS – PART 1

Chair: Robert Harley

11:40 am The results of EPAct/V2/E-89 study and data analysis – summary, *Aron Butler, Environmental Protection Agency*

12:15 pm **Lunch (Salon D)**

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[5] Planning Committee: Allen Robinson, Robert Harley, Maria Costantini, and Rashid Shaikh

FUEL COMPOSITION, ENGINE TECHNOLOGY, AND PM EMISSIONS – PART 2

Chair: Allen Robinson

1:15 pm Effects of fuel properties on particle emissions from GDI vehicles: Role of the PM Index, *Imad Khalek, Southwest Research Institute*

1:40 pm Fuel and after-treatment effects on particulate and toxic emissions from GDI and PFI vehicles: a summary of CE-CERT research, *Georgios Karavalakis, University of California at Riverside CE-CERT*

2:05 pm Characterization of GDI particle emissions during start-stop operation with alcohol fuel blends, *John Storey, Oak Ridge National Laboratory*

2:30 pm **Break**

2:45 pm Fuel effects on regulated emissions from modern gasoline vehicles, *Heather Hamje, CONCAWE*

3:10 pm Black carbon particle emissions from GDI vehicles operating on different fuels, *Tak Chan, Environment and Climate Change Canada*

3:35 pm Relative toxicity of old and new technology heavy- and light-duty mobile source PM, *Jorn Herner, California Air Resources Board*

PANEL DISCUSSION: INTEGRATING RESEARCH AND POLICY ISSUES

` *Chair: Daniel Greenbaum*

4:00 pm Opening comments: *Kathryn Sargeant, EPA; Geoff Cooper, Renewable Fuel Association; Matti Maricq, Ford Motor Company; Daniel Short, Marathon Petroleum Corporation*

5:00 pm **Adjourn**